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Simulation of Differential Die-Away instrument's response to asymmetrically burned spent nuclear fuel

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Abstract: Previous simulation studies of Differential Die-Away (DDA) instrument's response to active interrogation of spent nuclear fuel from a pressurized water reactor (PWR) yielded promising results in terms of its capability to accurately measure or estimate basic spent fuel assembly (SFA) characteristics, such as multiplication, initial enrichment (IE) and burn-up (BU) as well as the total plutonium content. These studies were however performed only for a subset of idealized SFAs with a symmetric BU with respect to its longitudinal axis. Therefore, to complement the previous results, additional simulations have been performed of the DDA instrument's response to interrogation of asymmetrically burned spent nuclear fuel in order to determine whether detailed assay of SFAs from all 4 sides will be necessary in real life applications or whether a cost and time saving single sided assay could be used to achieve results of similar quality as previously reported in case of symmetrically burned SFAs.

The results of this study suggest that DDA instrument response depends on the position of the individual neutron detectors and in fact can be split in two modes. The first mode, measured by the back detectors, is not significantly sensitive to the spatial distribution of fissile isotopes and neutron absorbers, but rather reflects the total amount of both contributors as in the cases of symmetrically burned SFAs. In contrary, the second mode, measured by the front detectors, yields a sizable sensitivity to the orientation of the asymmetrically burned SFA inside the assaying instrument. This study thus provides evidence that the DDA instrument can potentially be utilized as necessary in both ways, i.e. a quick determination of the average SFA characteristics in a single assay, as well as a more detailed characterization involving several DDA observables through assay of the SFA from all of its four sides that can possibly map the burnup distribution and/or identify diversion or replacement of pins.

Keywords: differential die-away, spent nuclear fuel, asymmetrical burn-up

1. Introduction

The Differential-Die Away (DDA) method is one of the techniques which is being investigated within the Next Generation Safeguards Initiative (NGSI) spent fuel project of the U.S. Department of Energy [1]. The main objectives of the NGSI spent fuel project are to develop and to test instrumentation for plutonium mass content determination inside commercially utilized spent fuel assemblies (SFAs), verify the operator's declarations in terms of the irradiation history parameters such as initial enrichment (IE), burn-up (BU), and cooling time (CT) and to test the SFA for partial defects which could signal a deliberate illicit diversion of nuclear material.

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Originally, 14 different non-destructive assay (NDA) techniques were chosen to be possibly applicable for spent fuel assay [1]. Following thorough simulations and evaluations, the DDA technique based instrument turned out to be highly promising and comprehensive method with a capability to measure multiplication of the assayed SFA [2], determine its fissile [3] as well as total Pu content [4], and quantify IE and BU [5]. Thus in the current, i.e. later, stages of the NGSi project, the DDA instrument research focuses on practical aspects of real life measurements and conditions of its deployment. Within this paper, we address the effects of asymmetric BU on the response of the DDA instrument, which, if significant enough, may dictate the final instrument design or force the assay of the SFA to be performed from all of its sides, thus potentially significantly increasing time and cost requirements of the measurement.

During its lifecycle, a fuel assembly in a commercial nuclear reactor utilized for electrical power production undergoes typically three or four cycles of irradiation [6]. After each cycle, its position within the reactor core is changed according to a sophisticated shuffling scheme dedicated to maximize the power production efficiency. As a result of such maneuvers, BU across the fuel assembly will not be constant since it reflects the conditions within the reactor at the positions where the fuel assembly was situated.

In particular, those fuel assemblies, which during their life cycle occupy one of the positions on the outer edge of the reactor core, may end up with a significantly asymmetric BU. As will be presented in later sections of this paper, under certain conditions, the differences in concentrations of isotopes that are being consumed or created during irradiations (be it fissile ^{235}U , ^{239}Pu or strong neutron absorbing fission products such as ^{155}Gd) may vary even by tens of percent from side to side of the fuel assembly[7]. Therefore the primary objective of this work was to investigate, by means of simulations, how an asymmetric BU across a nuclear spent fuel assembly (SFA) influences the outcomes of an assay performed with a DDA instrument.

2. The Differential Die-Away technique

The DDA technique is an active nondestructive assay technique which uses a short ($\sim 10^3\text{s} - 100^3\text{s}$ of μs) external neutron pulse from a neutron generator (NG) to deliver fast neutrons, which penetrate into the SFA where they start fission reaction chains that essentially assay the entire SFA. The neutrons from these induced fission reaction chains are detected using a set of neutron detectors surrounding the SFA, and their detection time distribution is analyzed to deduce information about its properties.

The typical distribution of times when neutrons are detected, following an interrogation pulse of $10\text{ }\mu\text{s}$, peaks around $20\text{ }\mu\text{s}$ and then falls off approximately exponentially. The dominant part of the DDA signal in these very early time domains ($<100\text{ }\mu\text{s}$) consists, however, of “burst neutrons”, i.e. neutrons that do not cause any fission and reach the neutron detectors either straight from the neutron generator or with only minimal scattering. In absolute terms, the contribution of burst neutrons is almost independent of the characteristic parameters of the assayed SFA and can be subtracted as a constant background as shown in [2]. The remaining part of the DDA signal, in the form of the distribution of neutron detection times, peaks typically between 50 and $70\text{ }\mu\text{s}$ after the interrogation pulse, and then falls off with a die-away constant that is initially ruled by the characteristic parameters of the SFA but later by the overall multiplication of the system.

Die-away time is therefore one of the most significant parameters of the instrument’s response. It has been traditionally defined as a neutron mean lifetime in a certain environment [8]. However, in our case of SFAs with a rather large multiplication, the die-away time reflects the mean lifetime of an entire neutron population that consists of multiple neutron generations which are simultaneously created and die

away as rather long fission chains develop and progress through the instruments various materials. Should the interrogated item contain no fissile material, the injected neutron population will die away quickly reflected by fast decrease in neutron count rate registered by the detectors. If any fissile material is present inside the assayed item, the life of the neutron population is extended by birth of new neutron generations by induced fission, resulting in a slower decrease of observed neutron count rates, i.e. longer die-away time. In contrast to fissile isotopes, the isotopes with high cross sections for neutron absorption shorten the life time of the neutrons, thus shortening the die-away time and changing both the shape as well as the magnitude of the distribution of times when the neutrons are detected [5]. In case of the DDA instrument, the die-away time is thus an implicit indicator of the balance between the neutron-producing fissile material and neutron-absorbing fission products and actinides.

In traditional passive neutron counting based techniques the neutron population evolution in time can be approximated by a single exponential, with the decay constant being the die-away time [8]. In the case of DDA, the neutron population evolution is more complicated, since the neutron spatial as well as energy distribution undergoes dramatic changes on the way from the NG to the SFA and then out to the individual detectors. Therefore, since it cannot be approximated by a single exponential, when a detection time distribution is obtained, an analysis of the DDA signal is performed in limited time domains where such approximation can be justified. Our attention was primarily focused on the time domain of 100-200 μ s, which in previous studies [2, 5] has been identified as the most promising time domain to extract unique information about SFA multiplication, IE, and BU.

Apart from the die-away time determined (if possible) for each time domain, the overall number of neutrons detected within each particular time domain is called the DDA signal and is generally related to the absolute amount and type of fissile material in the SFA. An example of a modeled detection time distribution of neutrons detected by all individual detectors of the DDA instrument is illustrated in Fig. 1. A pressurized water reactor (PWR) SFA with 4% IE, 15 GWd/tU BU, and 5 years CT has been chosen to demonstrate differences between the detection time distributions of burst and fission neutrons, and their sum. In this particular case, a burst neutron distribution reaches its maximum shortly after the end of the interrogating neutron pulse (~ 20 μ s) while the distribution of fission neutrons detection times peaks around 60 μ s. The total DDA signal reaches its maximum and levels off at around 30 μ s, and then decreases quasi-exponentially. While the fractions of detected burst and fission neutrons are about the same at ~ 40 μ s, the fraction of detected fission neutrons increases to $\sim 90\%$ at ~ 120 μ s and rises above 99% after ~ 220 μ s since the beginning of the neutron interrogation pulse.

Another important quantity that is characteristic of each SFA and can be measured by the DDA technique is *multiplication* (M) [2]. When defined as the number of neutrons produced in the SFA and the surrounding setup per incoming neutron from the neutron generator, it is referred to as “active multiplication”. This is however linearly related to “passive multiplication” that is defined as the number of neutrons produced in the SFA per neutron originating inside the SFA, primarily in spontaneous fission. In general, the multiplication, be it passive or active, is closely related to the BU of the SFA. Fresh fuel or fuel with low BU has typically a high fissile content and is thus characterized by high multiplication in contrary to the highly burned fuel with a low multiplication factor. Additionally, as the isotopic composition that defines the multiplication depends on IE and CT, the multiplication itself is dependent on IE and CT. In general, the average multiplication of a SFA is implicitly defined by all these parameters and tends to increase with higher IE, while decreases with higher BU and longer CT. In this study, a net multiplication calculated in MCNPX corresponds to the “active multiplication” described above, however, for simplicity will be referred to only as “multiplication”.

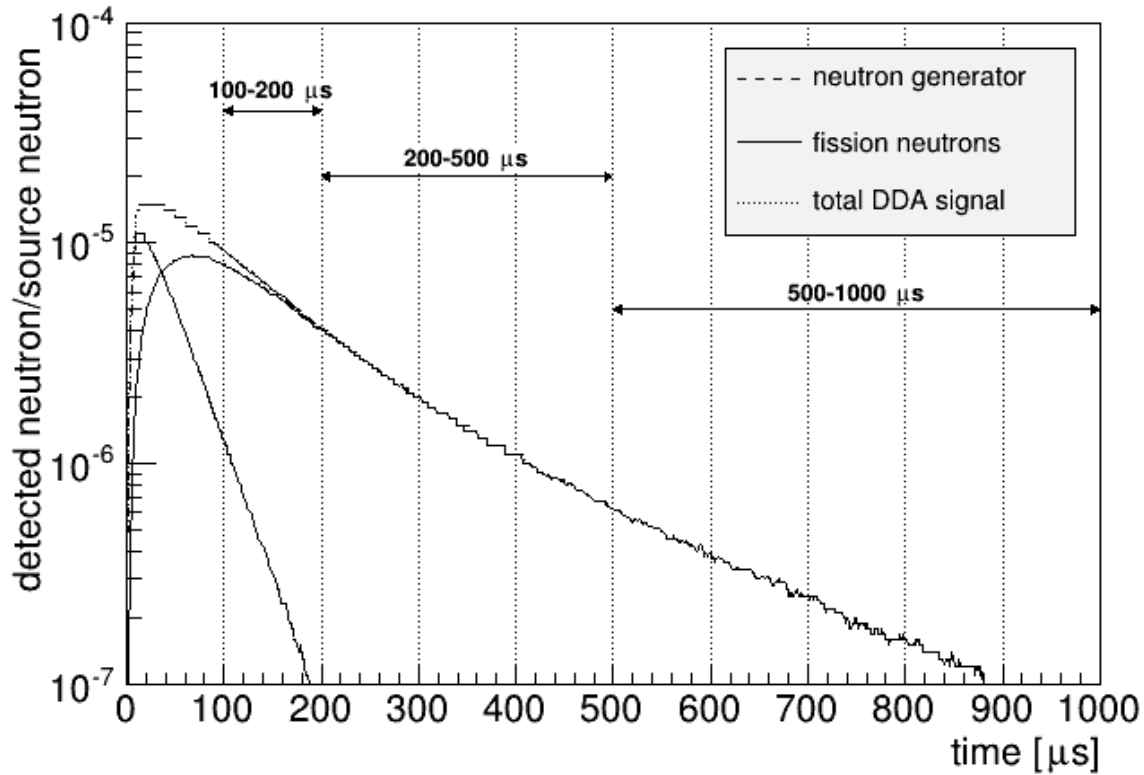


Figure 1: Simulated distribution of neutron detection times as registered by all DDA detectors for a 4 % IE, 15GWd/tU BU, 5 y CT PWR SFA. The evolution of total neutron signal (dotted line), fission neutron signal (solid line) and burst neutron signal (dashed line) is shown 0-1000 μ s after the start of the neutron interrogation pulse. Three different time windows (100-200 μ s, 200-500 μ s and 500-1000 μ s) typically utilized in the analysis are also displayed.

3. The Differential Die-Away Instrument

In this work, the use of a deuterium-tritium (DT) generator emitting neutrons with an initial energy of 14.1 MeV is assumed [9]. As the neutrons emitted from the NG slow down and their energies drop into the thermal and epithermal regions, they induce a significant amount of fission reactions in nuclei of fissile isotopes (primarily ^{235}U , ^{239}Pu and ^{241}Pu) often starting a fission reaction chain. The length of such chains depends on the overall isotopic composition of the SFA. Using ^3He detectors embedded in a high-density polyethylene (HDPE) moderator, which in turn is surrounded by a thin Cd liner, the instrument is essentially sensitive only to fast and epithermal neutrons, i.e. those coming from the NG or from induced fission before they have thermalized.

The design of the instrument simulated in this work is depicted in Fig. 2. It is a combined instrument for DDA and for delayed neutron (DN) detection. The latter technique utilizes two ^3He detectors, which are also encapsulated in HDPE but without a Cd liner, and the function of which is described more thoroughly in [3]. In order to simulate the neutron transport in the most realistic way, the DN detectors are included in the simulations performed in this work as well, with positions as indicated in Fig. 1. Their response is however not analyzed, since their proper function requires very different length and frequency

of interrogation pulse schemes and is a subject of other dedicated studies [10]. The DDA technique utilizes the remaining six detectors. In the analysis, the detectors are divided into two groups; the two detectors located closest to the NG at opposite sides of fuel assembly are referred to as front detectors, whereas the four detectors creating one group at the opposite side of the NG are referred to as back detectors.

The NG is surrounded by a tungsten block, with a primary function to tailor the energies of the source neutrons to sub-MeV energies [3]. Thus, while still fast and deeply penetrating, the first fission that occurs in the SFA is less likely to happen in ^{238}U nuclei, due to its relatively high energy threshold for fission, but rather in nuclei of fissile isotopes such as ^{235}U , ^{239}Pu and ^{241}Pu , which are of primary interest in the assay [11]. The tungsten block is enclosed by stainless steel, which is designed to reflect neutrons back towards the SFA, essentially to increase the efficiency of the instrument. Furthermore, a lead collar surrounds the SFA from three sides in order to significantly reduce the flux in the ^3He detectors from gamma rays emitted by the SFA.

The NG was modelled in Monte Carlo N-Particle eXtended (MCNPX) [12] as a voided 9.8 cm high cylinder with a 4.8 cm diameter, covered by a 0.1 cm thick layer of stainless steel. A point source was simulated in the center of this volume generating neutrons with an initial energy of 14.1 MeV, approximating the standardized NG based on the deuterium-tritium fusion reaction.

Standard ^3He detectors that are 2 inches in length, $\frac{3}{4}$ inch in diameter and filled by ^3He gas with 4 atm pressure were modelled in this study. The inner part of the detector was modelled as a 4.92 cm high cylinder with a 1.73 cm diameter covered by 0.08 cm thick layer of Al and filled by pure ^3He gas. This part was embedded in rectangular cuboid filled by HDPE covered by 0.1 cm thick layer of Cd in case of the 6 DDA detectors.

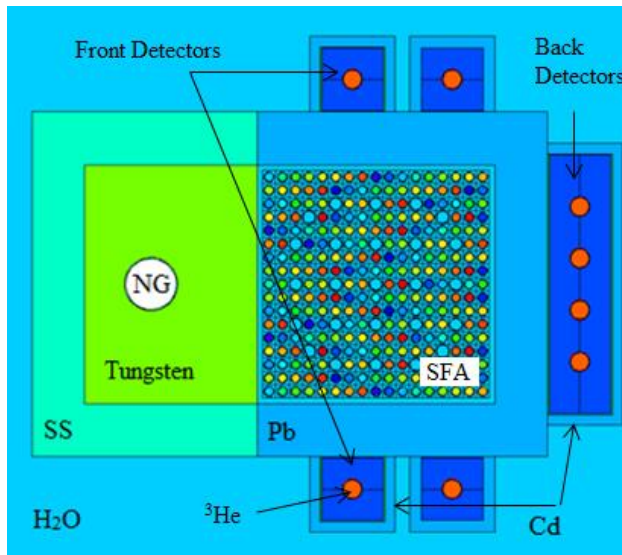


Figure 2: Schematic cross-sectional view of the DDA instrument with inserted SFA (top view) with the NG, SFA and detectors shown.

4. Modelling and Simulations

4.1. NGSF Spent fuel libraries

Within the NGS project a number of spent fuel libraries (SFL) with different SFAs for use in the MCNP code have been created. Typically, each SFL contains ensembles of SFAs with varying general parameters such as IE, BU, and CT and/or with varying BU history as defined by the position of the SFA in the nuclear reactor, density, temperature and chemical composition of the moderator. In this work two subsets of SFL2 were used (more details on SFL2 can be found in [13]). Both of these consist of PWR 17x17 standardized “Westinghouse”[14] fuel assembly models in form of the MCNPX inputs. Three different shuffling schemes (describing how the fuel moves in the reactor core during operation) were used to build the models of these SFAs.

In the simulations creating the SFLs, each SFA started as a fresh fuel assembly with specific IE (2, 3, 4, and 5 %) and was subsequently inserted into different positions inside the 1/8 core model of a PWR. The irradiation of the fuel assemblies was performed in up to three cycles, with each cycle resulting in an additional BU of 15 GWd/tU. In between the irradiation cycles, the SFAs were shuffled following one of three different schemes.

- Shuffling scheme #1 - the fresh fuel is inserted in the middle of 1/8 reactor core model before the first cycle, then moved closer to the core center before the second irradiation cycle and moved to the core periphery for the third irradiation cycle.
- Shuffling scheme #2 - the fresh fuel is first inserted in the reactor core center and subsequently moved outwards to the reactor core edge.
- Shuffling scheme #3 - inverse to the scheme #2; the fresh fuel is first loaded in the reactor core periphery, and then moved inwards for the second and third irradiation.

Shuffling scheme #1 concerns a shuffling pattern that agrees reasonably well with many commercial power producing reactors today. Shuffling scheme #3 resembles shuffling patterns of commercial reactors where fresh fuel is initially inserted in the periphery region, but it is often avoided to place the fresh fuel in the most peripheral positions. Shuffling scheme #2 is the most unrealistic one of the three schemes, partly because it makes it difficult to achieve an even power distribution in the core.

Part of SFL2, often referred to as SFL2a, contains fuels shuffled by scheme #1 with starting IE of 2, 3, 4, and 5 %. Fuels shuffled by scheme #2 and #3 are from a different part of SFL2, commonly referred to as SFL2c, but unlike the SFL2a, the SFL2c contains only fuels with 4% IE. One may note that the use of idealized SFA's and selected shuffling schemes is highly useful in learning about the instrument response. The results from this study should not be used as an estimate of absolute isotope concentrations of actual SFA's; this may be a topic for future studies.

All the shuffling schemes listed above create asymmetric distributions of fissile isotopes (with respect to its longitudinal axis) as well as of fission products and other neutron absorbers. However, such asymmetries become significant primarily in SFAs which occupy positions at the edge of the reactor core during the irradiation cycle. Therefore, the strongest asymmetries are formed in the third irradiation cycle (45 GWd/tU) for shuffling schemes #1 and #2; and in the first irradiation cycle (15 GWd/tU) for shuffling scheme #3. Examples of such asymmetric isotopic distributions are illustrated in Fig. 3 where the mass content in each fuel pin for the isotopes of ^{235}U , ^{239}Pu and ^{155}Gd is plotted for the case of PWR SFA with 4% IE, 15 GWd/tU BU, and 5 y CT which was irradiated as part of the shuffling scheme #3 at the edge of the reactor core. The mass of the isotopes inside the individual fuel pins is illustrated in Fig.3 in form of a 3D surface plot. We can see in the left panel of Fig. 3 how ^{235}U is depleted on the side facing the reactor core where the local burn-up is the highest. That is also the side where, due to the more intense burn-up, ^{239}Pu (center panel) and ^{155}Gd (right panel) get accumulated. In case of ^{235}U the side-to-side difference

may be only around 15%, but in case of ^{239}Pu and ^{155}Gd such differences may be even up to 40% and 20%, respectively.

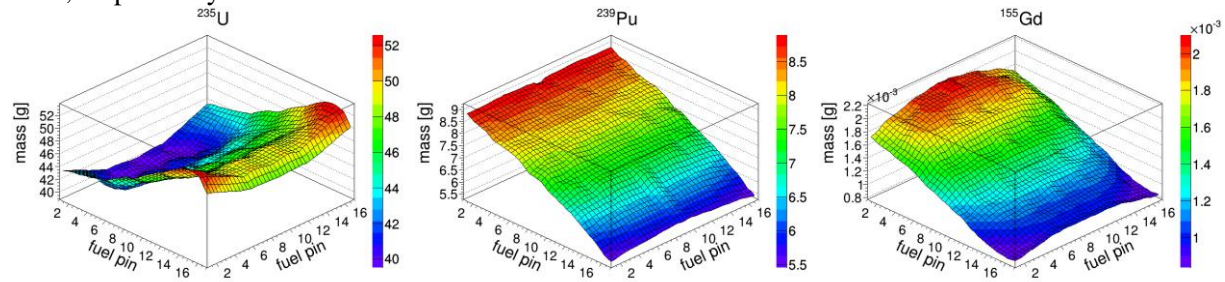


Figure 3: Asymmetric distribution of a ^{235}U (left panel), ^{239}Pu (middle panel), and ^{155}Gd (right panel) created by shuffling sequence #3 for a 4 % IE, 15Gwd/tU BU, 5 y CT SFA. (Note: 2D extrapolation of masses inside individual fuel pins was used to create the continuous surface plot)

4.2. Simulation of DDA Instrument Response

To study the sensitivity of the DDA instrument's response to the assay of SFA with asymmetric burn-up we used the MCNPX code to simulate the interrogation of the SFA from all of its four sides. The detector response was simulated following a 10 μs neutron interrogation pulse delivered by an external neutron generator. The geometry of DDA set-up as modeled in MCNPX is depicted in Fig. 2.

Given computer power constraints, the pulse produced by the NG was simulated to have $5 \cdot 10^8$ neutrons emitted in the time interval of 10 μs which represents only about 1-5 seconds of foreseen real life operation. However, even with such limitations, statistically significant results could be achieved. The neutrons were emitted isotropically with a uniform probability density function. The detection was modelled with a time dependent tally counting the number of neutrons detected in ^3He detector volume via the $^3\text{He}(n,p)^3\text{H}$ reaction [8] and its associated cross section during the time interval of 0-1000 μs after the start of the NG pulse.

The number of detected neutrons originating from fission reactions inside the SFA is directly related to the total fissile content inside assayed SFA. Nevertheless, another group of neutrons - the burst neutrons coming directly from the NG without undergoing any fission reaction - also contributes to the neutron count rate inside the detectors, especially in the early time domains after the pulse (~ 0 -200 μs). The burst neutrons were evaluated separately during the simulations and afterwards subtracted from the total count rate. This separation is enabled by using a function of MCNPX called *first fission*, which labels the detected neutrons into several groups depending on their origin. With this feature, it is possible to distinguish whether a neutron has been involved in a (n,f) reaction, and if so, the information on which isotope the first fission occurred on can be saved. It should be noted that this special feature is not included in publicly released versions of MCNPX, but it is an integral part of the recently released MCNP6 [15].

Each of the ^3He detectors was tallied separately in MCNPX. For the analysis of a total detector response, the contributions from all 6 detectors (2 front detectors, 4 back detectors) were summed in post processing. However, for the purposes of this study the front and the back detectors could be analyzed separately.

4.3. Previous studies on asymmetrically burned SFA

The SFAs with asymmetric BU distribution have been previously studied using different NGS techniques. The delayed neutron method, which uses the same detection design as DDA (Fig.2) has been investigated in work of Trellue et al. [10] for a sensitivity of the detection instrument to the different orientations of asymmetrically burned SFAs. The same set of SFA models has been used in [10] as in our work. Delayed neutron count rates summed up from all detectors did not indicate differences larger than 1-2 % when the SFA is oriented with different sides towards to the NG. These small differences were observed even for the most extreme case in BU asymmetry, a SFA from SFL2c, shuffling scheme #3 with 4% IE, 15 GWd/tU BU. While the delayed neutron fractions emitted from different positions of the SFA by ^{241}Pu , ^{239}Pu and ^{235}U differ significantly, the non-uniformity in the delayed neutron emission in the SFA was found to be compensated by the multiplication. Thus, results obtained in [10] were interpreted as not being able to reflect extreme asymmetry in BU distribution and were considered as a demonstration of the overall insensitivity of the detection instrument to the orientation of asymmetrically burned SFA inside the DN instrument.

In another study [7] passive gamma ray spectra were simulated for a High Purity Germanium (HPGe) detector collimated to observe a 5 cm section centered on the side of the SFA. Variations in simulated ^{137}Cs and ^{134}Cs peak intensities between different SFA sides presented in that work range between 20-40 % in case of 15 GWd/tU BU SFAs, characterized by the greatest gradient in BU. For the rest of the SFAs the gamma ray intensities for different sides vary only in the range of 3 to 5 %. Detected gamma rays were tracked and found to originate primarily from isotopes located in the most outer rows of fuel pins in the SFA, and they depend primarily on the number of fissions which take place in these particular fuel pins. Therefore, this instrument is considered to be dependent on orientation of the assayed SFA should it have an asymmetric burn-up.

5. Results

The work presented in this paper focuses on two subset of SFAs from SFL2a and SFL2c [13]; all with the same IE of 4% but varying BU of 15, 30, and 45 GWd/tU, and CT of 5 and 20 years. These SFAs underwent a simulated BU in a nuclear reactor according to three different shuffling schemes (in detail described in section 4.1. and [13]) resulting in 18 SFAs with different isotopic composition and spatial distribution. Additionally, for each of these 18 SFAs we simulated 4 different assembly orientations inside the DDA instrument in order to test the sensitivity of the DDA response to the asymmetric distribution of BU across the SFA assembly. Table 1 summarizes characteristic parameters of SFA's and the associated shuffling schemes used in this study.

Table 1: Characteristic parameters of SFAs, the associated shuffling scheme under which their burn-up was simulated and their attribution to the spent fuel libraries.

Spent Fuel Library	SFL2a	SFL2c	
Shuffling scheme	#1	#2	#3
IE [%]	4	4	4
BU [GWd/tU]	15, 30, 45	15, 30, 45	15, 30, 45
CT [year]	5, 20	5, 20	5, 20

The analysis of the simulated DDA response was performed in discrete time domains, all together covering the full dynamic evolution of the DDA signal after the interrogating neutron pulse. The choice of the time domains is identical to our previous work [2], i.e. 0-50 μs , 50-100 μs , 100-200 μs , 200-500 μs ,

and 500-1000 μ s. The die-away time was then obtained in time domains 100-200 μ s, 200-500 μ s, and 500-1000 μ s by a least squares fit of an exponential to the neutron detection time distribution. However, since the results for individual time domains lead to qualitatively the same conclusions, only results for the time domain of 100-200 μ s are presented here.

In Fig. 4, the DDA signals for all four orientations of each SFA relative to their four-value average are displayed as a function of the SFA multiplication. The left panel displays the differences in DDA signal as simulated for the front detectors, while the right panel displays the same quantity for the back detectors. As the multiplication tends to decrease with higher BU, the grouping of the points along the x-axis is indicative of various discrete levels of BU, in particular of 15, 30 and 45 GWd/tU.

When comparing data from the left (front detectors) and right (back detectors) panels of Fig.4, the apparent difference is the level of dispersion among the points corresponding to a single SFA, which differ only by the SFA orientation in the DDA instrument during the simulated assay. Depending on the particular shuffling scheme and BU, the difference in DDA signal caused by the orientation of the SFA may reach up to $\pm 3.3\%$. The two most extreme cases where the asymmetry is reflected by the largest spread in data points are observable for the SFAs of 15 GWd/tU BU shuffled by scheme #3, which was irradiated only once at the reactor core edge; and for the SFAs with 45 GWd/tU BU shuffled by scheme #2, which ended in the third cycle of irradiation at the reactor core edge. Should we follow the evolution of the BU asymmetry under the shuffling scheme #3, it can be found that with the SFA being moved from the edge towards the center of the nuclear reactor core, the BU asymmetry tends to disappear as can be observed by the decreased spread of the corresponding points in the left panel of Fig.4. For shuffling schemes #1 and #2 the trend is rather opposite, since the SFAs start their burn campaign in the middle of the reactor core and with higher BU are shuffled towards its edge where most of the asymmetric BU is developed.

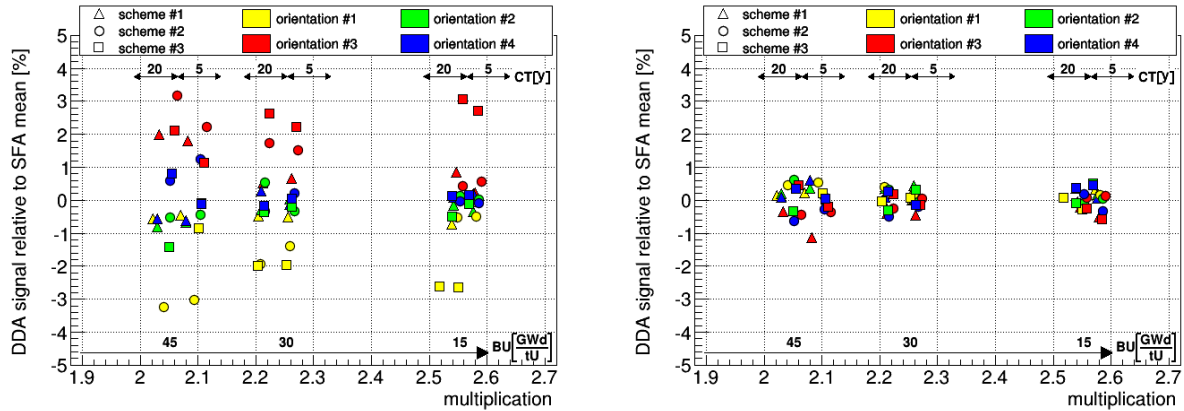


Figure 4: Differences in 4 simulated DDA signals of a particular SFA relative to its arithmetic mean displayed as a function of multiplication. Data points are illustrated separately for front (left panel) and back (right panel) detectors for 3 different shuffling schemes (#1: triangles, #2: dots, #3: squares). The approximate correlation between BU, CT and multiplication (M) is indicated by top, bottom secondary x axis, respectively. Different colors indicate various orientations of SFA.

However, in contrast to the sensitivity of the front detectors to the asymmetric BU, the back detectors do not seem to yield any such sensitivity as can be concluded from a very low and rather random spread of points in right panel of Fig.4, with the standard deviation being $\sim 0.2\%$. In order to interpret such apparent discrepancy, we need to consider the origin and the most probable path of the fission neutrons that are detected by individual detectors. As an illustration, Fig.5 displays a simplified schematic of neutron paths

on their way from the NG towards front and back detectors for a SFA in two orientations, 180° shifted. In the illustration, the SFA was conceptually divided into two equally sized regions representing volumes of low and high BU. As can be seen from both panels, depending on the orientation of the SFA, the neutrons that are detected by the front detectors need to pass only through the region of a rather similar BU, be it low in the left panel or high in the right panel. Depending on the local multiplication in the individual regions that trends with BU, the resulting signal in the front detectors will be either suppressed (high BU) or enhanced (low BU). The neutrons that are detected in the back detectors mostly also originated in their proximity, i.e. in region of low or high BU depending on the orientation of the SFA, but these neutrons are a product of a fission chain that was started by a neutron from the NG and as such had to traverse through the entire SFA, i.e. through both regions of different BU. Depending on the orientation of the SFA, the fission chain was first multiplied according to the multiplication of the region closer to the NG, then according to the multiplication of the region closer to the back detector. In the end, the total multiplication is about the same, no matter which BU region was encountered by the fission chain first, resulting in the insensitivity of the back detectors to the asymmetric BU.

As has been shown in previous work [5], in the case of a 100-200 µs time domain and SFAs with constant IE, the die-away time also depends linearly on the multiplication. It is thus natural to expect certain sensitivity in the die-away time measured by the front detectors on the orientation of the SFA as well. After all, the up to the 3.3% difference in the DDA signal, as seen in left panel of Fig.4, may be interpreted also as a change in multiplication perceived by the front detectors. Left and right panels of Fig.6 display relative changes of the die-away time of the simulated DDA signal in the time domain of 100-200 µs with respect to the average value calculated from all four different orientations of a particular SFA inside the DDA instrument. In both of these plots, however, the spread of the points seems to be rather constant at a level of $\sim \pm 1\text{-}2\%$ and also random with respect to the orientation of the SFA. There seems to be no apparent sensitivity of the die-away time to the asymmetry of the BU which appears to be in contradiction to our expectations.

In order to understand the lack of the observed sensitivity, we need to evaluate individual terms of the linear relationship between the multiplication and die-away time τ as shown in Eq. (1):

$$\tau[\mu\text{s}] = a \cdot M + b \quad (1)$$

where the multiplication M is in the form of DDA signal in the time domain of 100-200 µs; a and b are constants characterizing the specific instrument design and the investigated SFA [5]. In case of the 4% IE SFAs, the numerical value of b is ~ 60 µs, while the value of the term $a \cdot M$ ranges from 10 to 50 µs depending on the BU and CT [5]. Therefore a 3% change in M as indicated by change in the DDA signal itself will result in only $\sim \pm 0.2\text{-}1.5\%$ change in τ . Such a small change is however comparable to the uncertainty of the exponential fit from which the die-away constant is determined, which is reflected by a nearly constant dispersion of the points in Fig. 6 regardless of the BU or position of the detectors. Thus while in principle the die-away time of the DDA signal should also reflect the asymmetry of the SFA BU, real-life limitations on achievable statistics may make its use impractical.

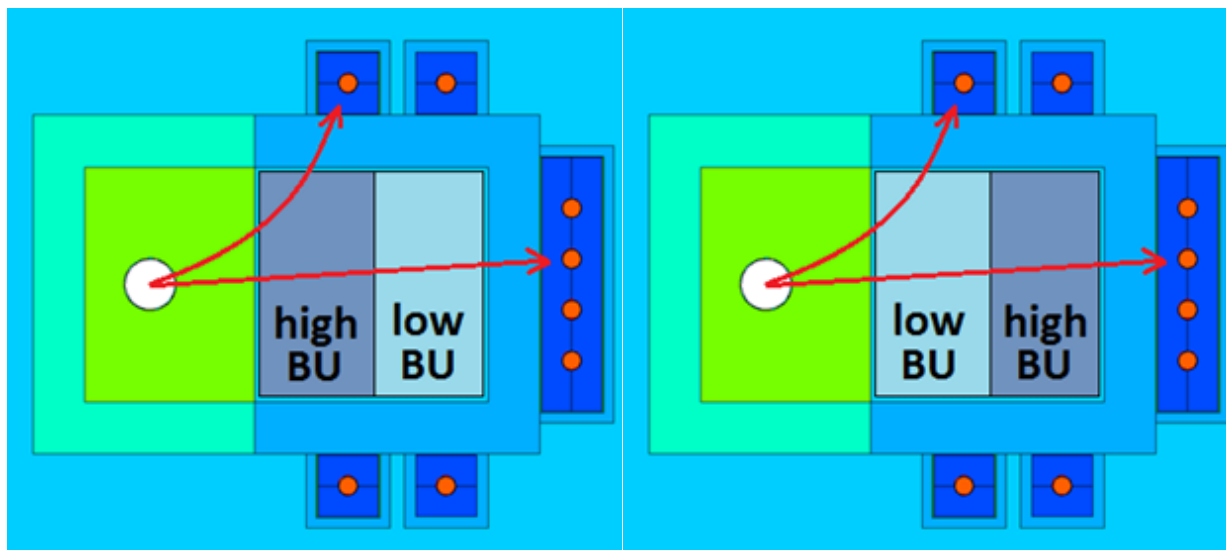


Figure 5: Schematic illustration of the most probable paths of neutrons detected in front and back detectors exemplified for two orientations of the SFA inside the DDA instrument (left panel: high BU in front part of SFA; right panel: low BU in front part of SFA).

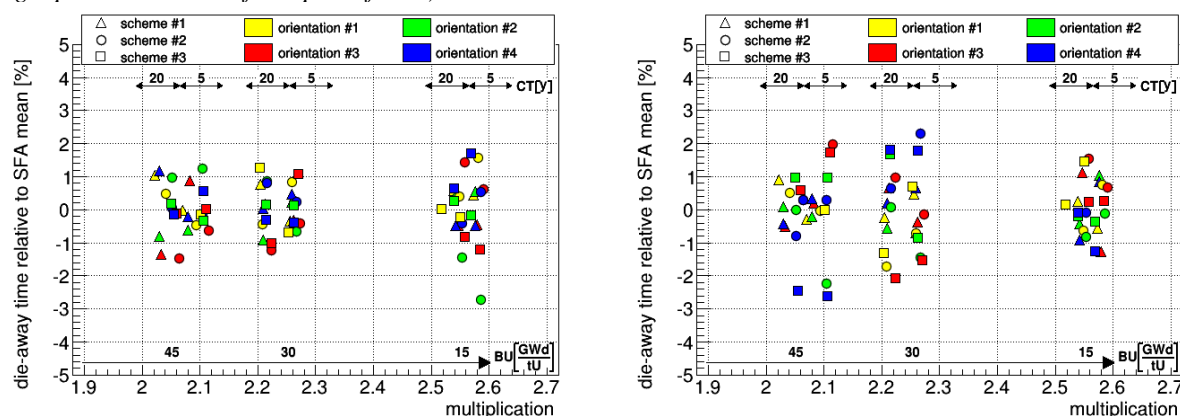


Figure 6: Differences in 4 die-away times of the simulated DDA instrument response obtained for 4 different orientations of each SFA expressed relative to its mean displayed as a function of multiplication. Results are presented separately for front (left panel) and back (right panel) detectors for 3 different shuffling schemes (#1: triangles, #2: dots, #3: squares). The approximate correlation between BU, CT and multiplication (M) is indicated by top, bottom secondary x axis, respectively. Different colors indicate various orientations of SFA.

6. Conclusions and Outlook

By means of simulations, we have demonstrated that an instrument based on the DDA technique, proposed within the NGSF spent fuel project, can be used for NDA assay of asymmetrically burned SFAs. Depending on the location of the individual neutron detectors with respect to the neutron generator and the spent fuel assembly, the DDA instrument can operate in two different modes, one being sensitive to the asymmetric burn-up, the other one not. This conclusion is based on a separate analysis of the instrument's front and back detector response to the interrogation of asymmetrically burned SFAs from all four different sides. The front detectors display a sensitivity to different fuel orientation, which in case of the most asymmetrically burned SFA (where e.g. the concentration of ^{239}Pu differs by up to 40% from

side to side) translates into $\pm 3.3\%$ change in the detected neutron rates. In contrast to that, the neutron rates measured simultaneously by the back detectors do not seem to yield any systematic change. Moreover, the standard deviation of $\sim 0.2\%$ of the signal measured by the back detectors can be attributed to expected statistical fluctuations.

Considering the difference in the quality of the signal measured by individual neutron detectors of the DDA instrument at different positions with respect to the assayed SFA, we conclude that the DDA instrument has a potential to be used under various operational demands, be it fast single-assay to determine mean values of multiplication, burn-up or total plutonium content, as well as a multi-side assay aiming to map spatial variation of different SFA properties in greater detail.

This project is organized in cooperation between Uppsala University and Los Alamos National Laboratory, with future possibility to perform testing measurements with the DDA instrument on a set of 25 PWR and 25 BWR SFAs varying in IE, BU, and CT parameter. Future studies will focus on the detailed optimization of the instrument design followed by manufacturing of such an instrument. The overall goal of this project is to develop and test methods, which will be applicable in the nearest future for the verification of Swedish spent fuel before its final encapsulation and deposition into the final repository.

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8. References

[1] S. J. Tobin, H. O. Menlove, M. T. Swinhoe, et.al.: “*Technical cross-cutting issues for the next generation safeguards initiative's spent fuel nondestructive assay project*”, Journal of Nuclear Materials Management Vol. 40, No. 3 (2012) pp 18-24.

[2] V. Henzl, M. T. Swinhoe, S. J. Tobin, and H. O. Menlove: “*Measurement of the Multiplication of a Spent Fuel Assembly with the Differential Die-Away Method within the Scope of the Next Generation Safeguards Initiative Spent Fuel Project*”, Journal of Nuclear Materials Management Vol. 40, No. 3 (2012) pp 61.

[3] P. C. Blanc, S. J. Tobin, S. Croft, and H. O. Menlove: “*Plutonium Mass Determination in Spent Fuel-Delayed Neutron Detection in an Integrated Delayed-Neutron and Differential Die-Away Instrument with ^3He Detectors and a DT Generator*”, Los Alamos National Laboratory report LA-UR-11-01912 (2011).

[4] V. Henzl, S. Croft, J. Richard, M. T. Swinhoe, and S. J. Tobin: “*Determination of the Plutonium Content in a Spent Fuel Assembly by Passive and Active Interrogation using a Differential Die-Away Instrument*”, Nuclear Instruments And Methods A 712, 83-92, 2013, doi:10.1016/j.nima.2013.02.006

[5] V. Henzl, M. T. Swinhoe, and S. J. Tobin: “*Direct Measurement of Initial Enrichment and Burn-up of Spent Fuel Assembly with a Differential Die-Away Technique Based Instrument*”, Proceedings of Institute of Nuclear Materials Management conference, Orlando, Florida, 2012

- [6] W. M. Stacey: *"Nuclear reactor physics 2nd ed., completely rev. and enlarged edition"*, Weinheim: Wiley-VCH, c2007, page 210-211, ISBN 978-3-527-40679-1
- [7] J. D. Galloway, S. J. Tobin, H. R. Trellue, and M. L. Fensin: *"The Role of Monte Carlo Burn-up Calculations in Quantifying Plutonium Mass in Spent Fuel Assemblies with Non-Destructive Assay"*, Budapest: European Safeguard Research and Development Association, 2011
- [8] D.Reilly, N. Ensslin, H. Smith, Jr., and S.Kreiner: *"Passive Nondestructive Assay of Nuclear Materials"*, ISBN: 0-16-032724-5
- [9] *"Sodern Neutron Generator Genie 16 GT"*, available online at: ["http://www.sodern.com/sites/docs_wsw/RUB_79/Genie35.pdf"](http://www.sodern.com/sites/docs_wsw/RUB_79/Genie35.pdf), [2014-11-24]
- [10] H. R. Trellue, A. B. McKinney, A. Favalli, S. J. Tobin, and T. L. Burr: *"Sensitivity Of the Delayed Neutron Technique to Variations in the Spent Fuel Libraries and Neutron Absorbers"*, Proceedings of Institute of Nuclear Materials Management conference, Orlando, Florida, 2012
- [11] R. B. Firestone, and V. S. Shirley: *"Table of Isotopes. 8th ed."*, New York: Wiley, c1996, ISBN 0471-14918-7.
- [12] J. F. Pelowitz (Editor), MCNPXTM User's Manual Version 2.7.0, Los Alamos National Laboratory Report LA-CP-11-00438, April 2011
- [13] H. R. Trellue, J. D. Galloway, N. A. Fischer, and S. J. Tobin: *"Advances in Spent Fuel Libraries"*, Proceedings of Institute of Nuclear Materials Management conference, Palm Desert, California, 2013, Los Alamos National Laboratory Report LA-CP-13-24074,
- [14] *"Westinghouse RFA-2 Design"*, available online at: ["http://westinghousenuclear.com/Portals/0/Operating%20Plant%20Services/Fuel/Fuel%20Products/NF-FE-0005%20RFA-2_PWR.pdf"](http://westinghousenuclear.com/Portals/0/Operating%20Plant%20Services/Fuel/Fuel%20Products/NF-FE-0005%20RFA-2_PWR.pdf), [2014-11-24]
- [15] J. F. Pelowitz (Editor), MCNP6TM User's Manual, Los Alamos National Laboratory Report LA-CP-11-01708, April 2013